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1. INTRODUCTION

Tritium, the radioactive isotope of hydrogen, is formed abundantly in nuclear reactors using heavy water as the moderator. A possible rupture or leak in water lines can result in a radiation health hazard by spreading significant quantities of tritiated water (HTO) or HTO vapor into the atmosphere. Accidents and poor handling procedures in industry and various research laboratories utilizing the isotope in high activities can also result in acute or chronic exposures, especially if HTO is involved. Of all the possible compounds that contain tritium, HTO requires the most precautions. Physically, it may appear that tritium is harmless due to a beta spectrum characterized by a maximum energy of 18 keV and an average energy of only 5.5 keV. The average beta range in water is only a few microns and can hardly be considered hazardous as an external source of radiation. However, tritium in the oxide state is readily absorbed by the lungs and skin and is assimilated into body fluids. The relative hazards presented by environments contaminated with HTO vapor in contrast with molecular tritiated hydrogen are demonstrated by the Maximum Permissible Concentrations (MPC) of 5×10^{-6} μCi/cc and 2×10^{-3} μCi/cc, respectively.

The need for adequate monitoring techniques has led to the investigation of many possible monitoring systems. The difficulties

associated with the detection of very low energy radiations have necessarily complicated basic detection devices, and individual specifications are, therefore, often necessary for each particular monitoring requirement. Particular applications require consideration of factors such as type of media to be monitored, desired sensitivity, possible effects of the surrounding environment, and a host of other equally important parameters before a successful tritium monitor can be designed, constructed, and proven useful. In short, almost any successful monitoring system can be regarded as an accomplishment.

Practical monitors can be separated essentially into two groups. One group consists of monitors that render constant or immediate indications of activities within the monitored media. The second includes tritium collection systems that utilize water vaper collection traps for subsequent analysis of tritium. The immediate readout feature of the first group and high sensitivity of the second may, in a given situation, make the use of one more attractive than the other, or may require the use of both.

2. CONSTANT READOUT MONITORS

The basic components of the first group, or constant readout monitors, are the detector and its associated electronics, readout and, if needed, a system that constantly exchanges the monitored medium. The heart of the system is the detector, usually consisting of an ionization chamber, proportional counter, or scintillator-photomultiplier (PM) tube combination. Since the ionization chamber

collects the ions produced by ionizing radiation a current amplifier comprises the associated electronics. In contrast, the proportional counter and PM tube are normally pulse devices and therefore require pulse handling electronics.

The readouts most often used for the various systems are meters that may be calibrated with either counts per minute, units of specific activity, increments of MPC, or other values that may be applicable. These units are all interrelated and may be converted using proper calibration constants. A second form of readout is the scaler that is used with pulse systems. This readout is not constant but the integration period can be made short and repetitive, thereby simulating a constant readout and simultaneously increasing sensitivity.

With three detection devices available, one may infer that each has its merits and, similarly, its limitations. A brief examination of features peculiar to the devices substantiates this inference.

2.1 The Ionization Chamber

The current available from the ionization chamber is simply the rate of ion collection from inside the chamber. These ions can be caused by any type of ionizing radiation of any energy; therefore, energy discrimination and the ability to partially distinguish the tritium component from components due to other radiations are impossible. Also, the current is usually extremely minute. A simple calculation indicates a maximum current of 4.8×10^{-15} (amp/liter)/MPC. The sensitivity of an ionization chamber is therefore dependent upon chamber volume and amplifier response. With currents this low a number

of seemingly insignificant factors may cause significant errors. Waters[1] of Johnston Laboratories, Inc., reports these factors to be cigarette smoke, aerosols and ions, moisure and condensation, ambient gamma-ray field changes, radon, clogged filters, fission product gases, absorption and memory effects, and instrument calibration factors. Thus, with so many precautions necessary for its use, a practical ionization monitor usually employs a filter and ion trap at the input. The use of gamma compensation by means of a similar but closed chamber that negates response to gamma is also beneficial.

As impractical as it may appear the ionization chamber is the most popular device for general tritium detection in air. The construction is relatively simple and the measurement of currents has been easier than the measurement of pulses. The chamber came into use earlier than the other detectors and has therefore become recognized as standard. It is also accurate if used judiciously.

As stated earlier the maximum current possible is only 4.8×10^{-15} amp for each MPC contained in one liter. The Kanne chamber (Figure 1) monitor which is used at the Atomic Energy Commission's Savannah River Plant illustrates the necessity for large chambers if high sensitivity is to be achieved. According to Morris [2] of this installation, two sizes of chambers, 51.8 liters and 18.5 liters, are used routinely. The absence of a gamma compensation device requires installation of the chamber in a low-background environment, but remote areas can be monitored by use of hoses. The sensitivities for the 51.5- and 18.5-liter chambers are 2×10^{-6} and 8×10^{-6} μCi/cc, respectively.

Various designs for gamma compensated ionization chambers can be found. Anthony[3] describes a compensated system that uses a rectangular-shaped sensitive chamber and a similar but closed chamber adjacently mounted for compensation. The device has overall dimensions of 5 x 10 x 16 inches and a reported sensitivity of 4×10^{-5} μCi/cc.

A similar detection device is the Pittendrigh and Vousden[4] chamber, consisting of two identical half-cylinder chambers coupled across the plane surface. These also are constructed so that one chamber is closed and serves as a compensation device.

A system with gamma compensation of a different design is that of Osborne and Cowper[5]. Because the spatial separation of chambers in previous designs has made them dependent on gamma field direction, the compensating chamber is placed inside the active chamber (figure 2) so that geometrical symmetry provides equal response in essentially all directions. This arrangement is used in an area monitor of 40-liter capacity and two smaller portable versions of 1.2-liter and 0.3-liter capacities. The most sensitive scale for the 40-liter chamber is 0-1.5 MPC, or 0-7.5 $\times 10^{-6}$ μCi/cc. For the smaller chambers a portable vibrating reed electrometer makes possible the corresponding scales of 0-10 MPC and 0-45 MPC for the 1.2-liter and 0.3-liter models, respectively. An interesting feature, however, is the inevitable change in compensation due to different ion collecting efficiencies for the electrodes.

2.2 The Proportional Counter

The proportional counter is similar in principle to the ionization chamber but differences in design and bias, and the requirement for a particular counting gas, changes the chamber into a pulse device. The

proportional relationship between the pulse height and the number of ions produced per absorbed ionizing particle allows the use of energy discrimination circuitry. This capability is significant in reducing monitor response to radiation other than the tritium beta. The necessity of keeping the counting gas intact requires premixing with the monitored gas in specified proportions. This process inhibits general use of the counter and continuous use at high sampling rates can be uneconomical. The sensitivities obtained with such counters, however, can be extremely high. An alternative method of designing a counter which is conservative in gas use is the thin window proportional counter. In this case the tritium beta must penetrate the thin film that forms part of the counter's walls. Procurement of sufficiently thin film, however, is a considerable hindrance.

The use of proportional counters for tritium monitoring was initiated by Driver,[6] who was able to detect easily 2×10^{-5} μCi/cc with a counter volume of only 43 cc. This sensitivity was later increased by Ehret[7] to 10^{-7} μCi/cc or 0.02 MPC using counters with less than one-liter volume. To obtain these sensitivities, commercial counters were used which employed complex electrode arrangements (Figure 3) which, when used with appropriate electronics, made energy discrimination, anticoincidence and gamma compensation possible. These counters however are of the flow-through type which may make them impractical for general monitoring.

A thin window proportional counter designed for general monitoring has been described by Block and associates.[8][9] This monitor, like the ionization chambers, employs two identical counters (Figure 4), one of which is used for gamma compensation. Because the thin

window over the compensating counter is opaque only to tritium beta, compensation occurs also for high energy beta and alpha. The response for this system is 577 cpm/MPC.

2.3 The Scintillator-PM Tube

The third type of detector used in tritium monitors is the scintillator-PM tube detector and it differs greatly from the previous detectors. In this detector, absorbed ionizing radiation causes the scintillator to emit photons of light. If detected by the PM-tube a pulse of current is generated. The proportional relationship applies also for this detector and use of energy discrimination circuitry is usually employed. An outstanding feature of this detector over the other two is its ability to monitor liquids directly. The mechanics associated with this device which determine its practicality are too complicated to state briefly but are essentially identical to those associated with the liquid scintillation counter.

The use of scintillators for tritium monitors has been necessarily delayed until the last few years. Compact electronics, high quality scintillators and PM-tubes have now, however, brought attention to their potential usefulness.

Because of the availability and convenience offered by the plastic scintillator, its use in tritium detectors has been explored more than any other scintillator. With the detector geometry shown in Figure 5, Sannes and Banville [10] reported a sensitivity of 0.2 MPC. The detector consisted of two strips of plastic scintillator rolled together to form a double spiral. By making use of the energy

discrimination circuit in the electronics, gamma compensation was performed. The entire system was designed to be battery powered; hence portability was accomplished.

A detector using 40 parallel 2 in. x 2 in. sheets of plastic scintillator was constructed at the EERL. It was tested with electronics equivalent to those in liquid scintillators. The sensitivity also was found to be 0.2 MPC but required use of lead to shield the detector from local gamma background. Static electricity was observed at times and memory effects from HTO were judged to be too severe. The plate detector was temporarily abandoned and the design of Figure 6 was investigated.[11] This detector, using a thin layer of anthracene coating on the rods, also exhibited a sensitivity of 0.2 MPC. The use of anthracene greatly reduced background sensitivity and no shielding was required in the local environment. The memory effects were also greatly reduced and the design allowed monitoring of water and filtered urine with a sensitivity of 10 cpm for each μCi/liter. Prolonged use of this detector proved, however, that anthracene did not make a suitable scintillator for a practical detector because of quenching from oxidation.

Other parallel plate detectors have been investigated by Prevo and Santomassimo[12] (Figure 7) and Osborne.[13] The latter's detector, consisting of sixty 5 cm x 5 cm plates, rendered impressive results. This detector gave a response for water of 24 cpm /(μCi/liter). For tritium in air the same detector exhibited the extremely high response of 150 cpm/(10^{-6} μCi/cc), or rather, 750 cpm/MPC.

Two other detection systems that should be mentioned are the tritium in air detector of Prevo and Santomassimo[14] and the automatic tritium-in-urine analyzer that was designed by Osborne.[15] The former system uses a single crystal of $CaF_2(Eu)$, a nonhygroscopic inorganic scintillator which does not suffer from memory effects commonly observed with other scintillators. The latter system automatically mixes aliquots of urine with liquid scintillator and analyzes the solution for tritium.

3. TRITIUM COLLECTION SYSTEMS

The second group of monitors which use water vapor collection traps may consist simply of an air pump, air volume metering device, and a single trap. A somewhat more complex system would be required however if accurate determinations are to be made for air containing different forms of tritium. The sensitivity of these systems depends primarily upon the volume of air sampled and the type trap used. The decision of what trap to use is determined by weighing the convenience, sensitivity, and accuracy afforded by the various traps. Air that is pumped through a desiccant such as Drierite[T.M.] and silica gel will lose approximately one hundred percent of its water vapor to the desiccant as long as saturation is not approached. The vapor is then retrieved by distillation. If a lower sensitivity is tolerable, the vapor can also be retrieved by mixing the desiccant with distilled water and letting the solution sit until displacement has occurred.

Traps often used, other than desiccants, are cooled condensation traps and water bubblers. The cooled condensation trap yields high

T.M. - Drierite is a trademark of the W. A. Hammond Drierite Company, Xenia, Ohio.

sensitivity since aliquots of the condensed vapor can be obtained without dilution. However, care must be exercised to ensure complete collection. If incomplete but constant collection can be tolerated the water bubbler may often prove to be satisfactory. Other investigators[10, 17, 18] have shown that collection efficiencies above ninety-five percent can be obtained with relatively small volumes of water. The bubbler does necessitate dilution but a lower sensitivity may not be considered a hindrance due to the convenience afforded.

An example of this second group that has been used successfully at EERL is shown in Figure 8. Sequential components in this particular system are: (1) particulate filter; (2) Drierite canister for removing water vapor; (3) combustion chamber for converting hydrocarbons to water; (4) Drierite canister; (5) air volume meter, and (6) air pump. The filter and Drierite canisters are analyzed in the lab after sample collection for accurate specific activity determinations.

4. CONCLUSION

The selection of the various units, μCi/cc, cpm/(μCi/cc) cpm/MPC, and MPC, used to indicate a system's ability to detect tritium was not deliberate. These were the units chosen by the various authors. Sensitivity should mean the threshold of activity at which an instrument begins giving a meaningful measurement. Unless the conditions under which the measurement takes place are stated, it must be assumed that the measurement is made under ideal conditions for obviously, in high gamma fields, any instrument's "good sensitivity" can be

destroyed. The system's response such as cpm/MPC, however remains essentially constant and is, therefore, more descriptive.

The systems described have illustrated only partially the various systems and techniques that can be used for tritium monitoring and meaningful contributions have been made by many other authors. A specific situation can require its own peculiar system of monitoring. The parameters that affect the final activity determinations must be taken into account and complete knowledge of a system's limitations is of extreme importance.

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Table 1. Characteristics of detectors used in tritium monitoring

	Ionization	Flow-through Proport. Counter	Thin Window Proport. Counter	Scintillation Counter
1. Sensitivity	Good	Good	Poor*	Good
2. Media Monitored	Gas	Gas	Gas	Gas/Liquid
3. Energy Discrimination	No	Yes	Yes	Yes
4. Gamma Compensation	Yes	Yes	Yes	Yes
5. Size (MPC Levels)	Large	Depends on Gas Supply	Small	Small
6. Ease of Shielding	Impossible	Fair	Better	Best
7. Ease of Operation	Poor	Poor	Good	Good
8. Availability	Comm. & Develop.	Comm. & Develop.	Developmental	Developmental
9. Portability	Poor	Poor	Good	Good
10. Reliability	?	?	?	?

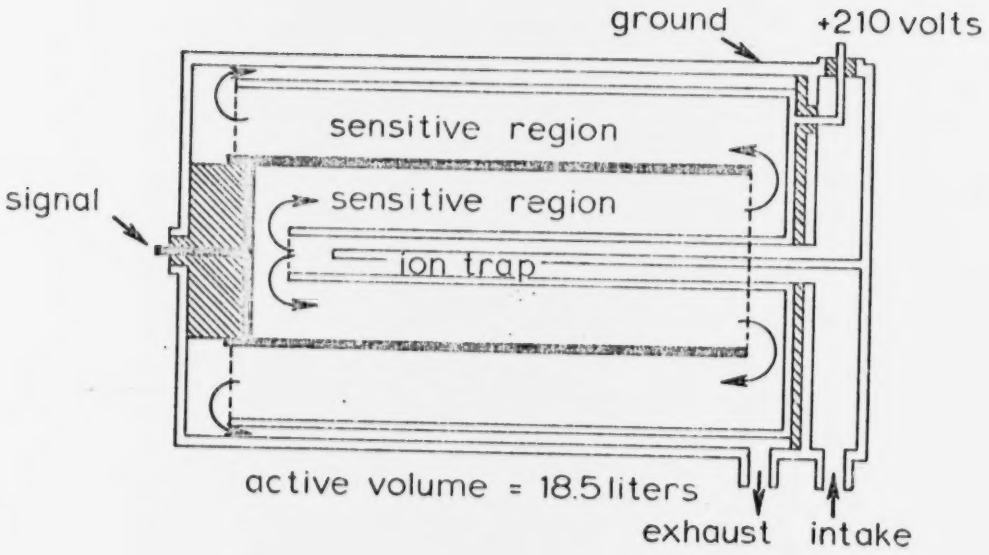

Figure 1 Kanne Chamber

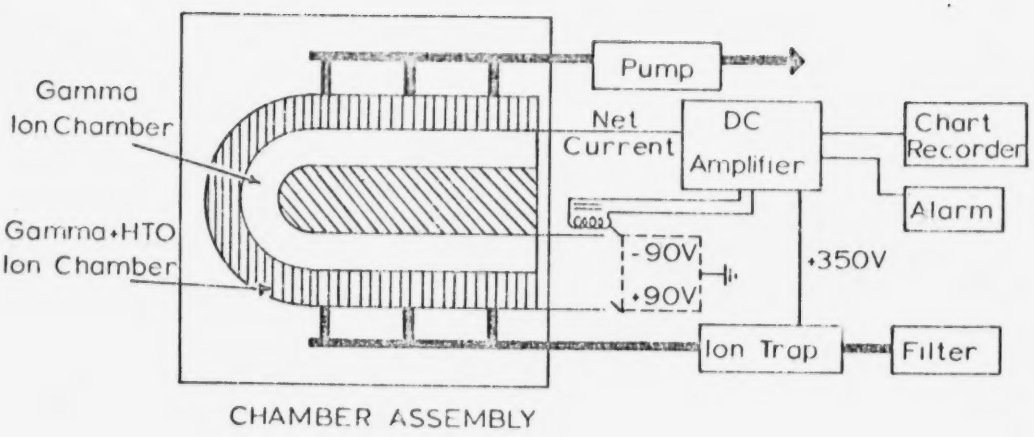

Figure 2. Osborne Tritium Monitor

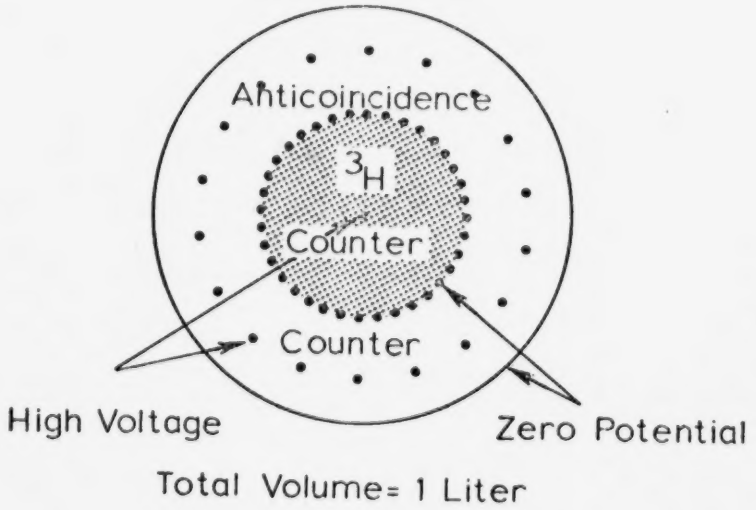

Figure 3.Multi-Electrode Proportional Counter

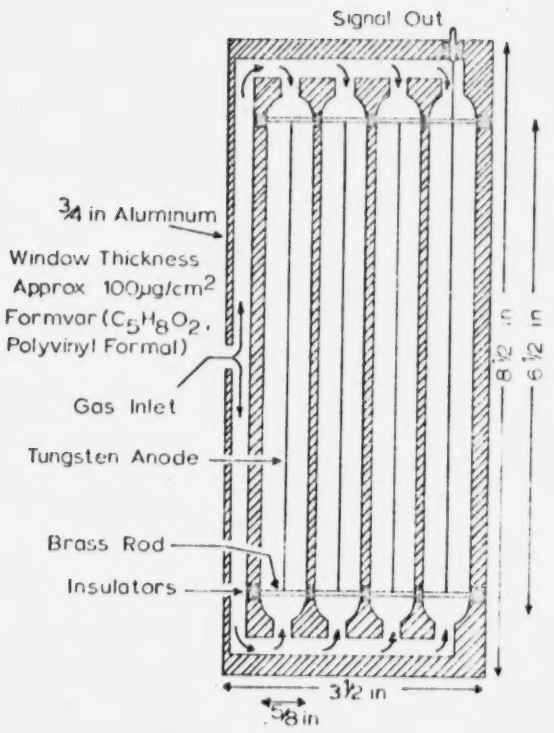

Figure 4. Internal View of Thin Window Proportional Counter

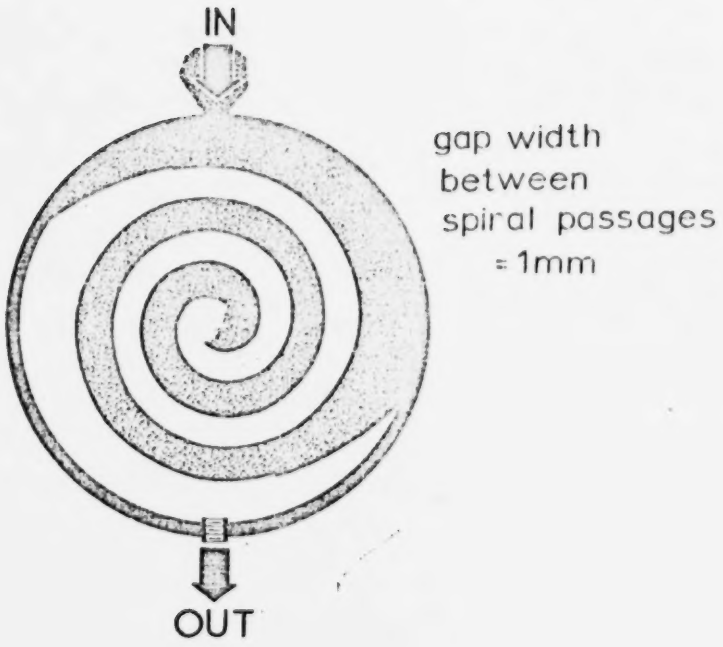

Figure 5. Spiral Scintillator Detector

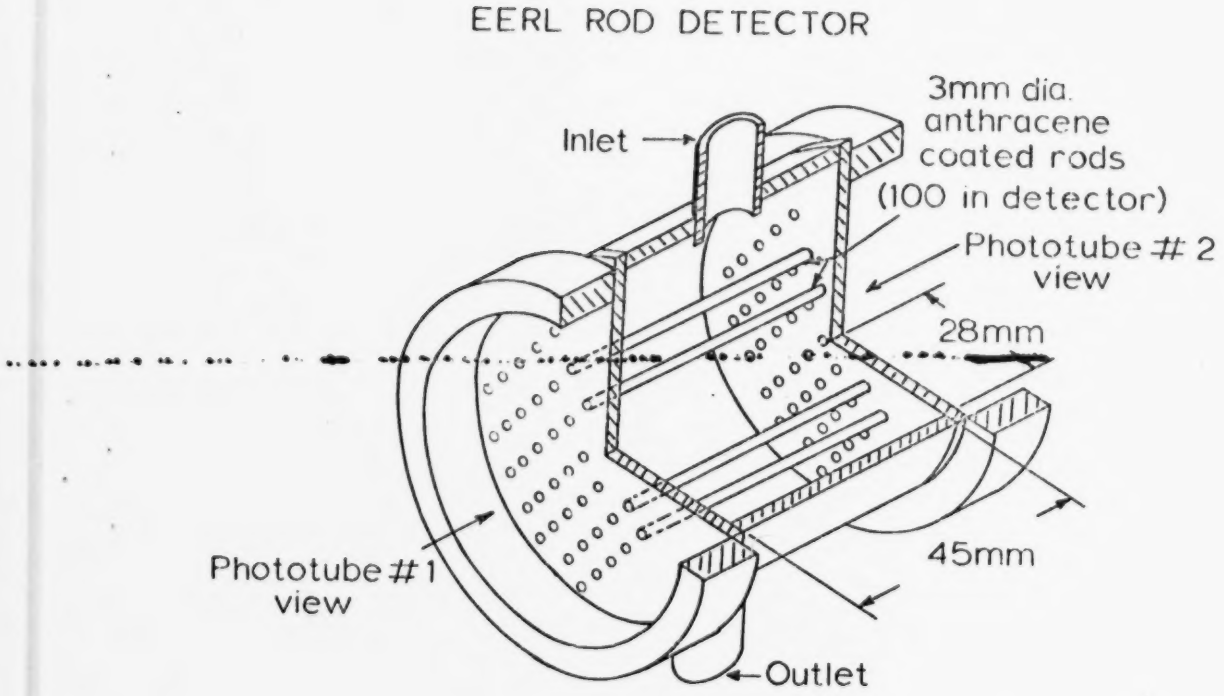

Figure 6. EERL Rod Detector

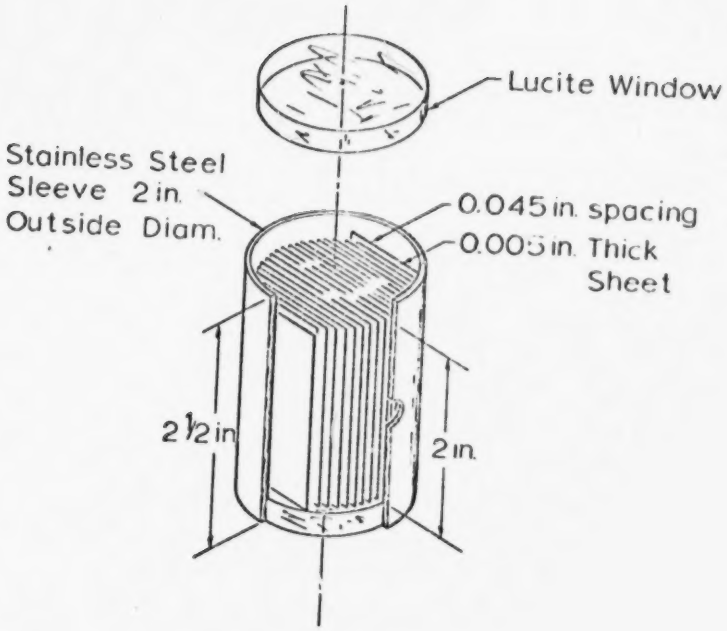

Figure 7. Parallel Plate Scintillator

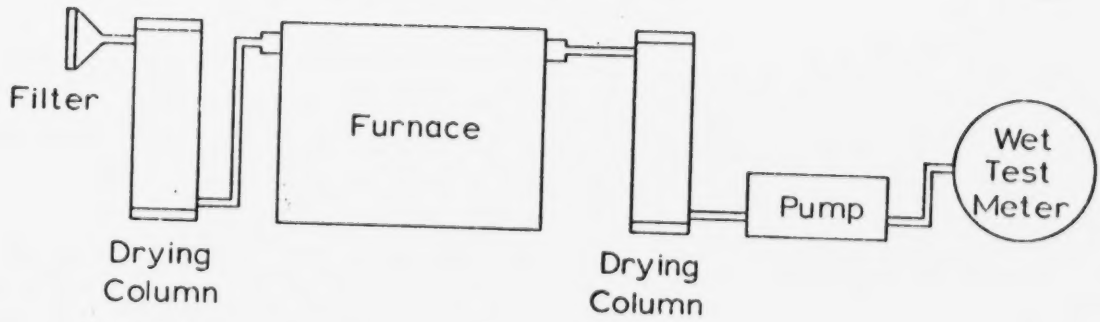

Figure 8. Combustion Train

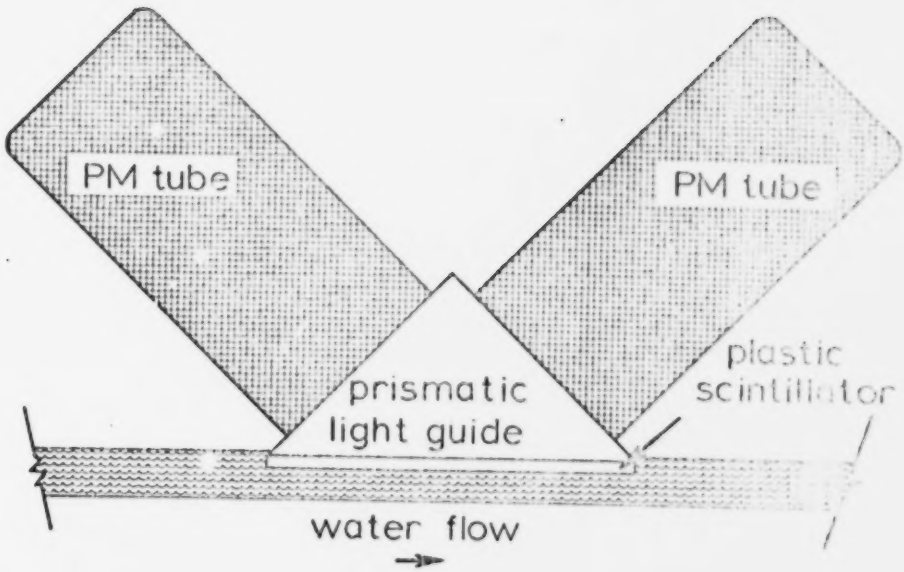

Figure 9. Continuous Water Monitor